Excitonic condensation of massless fermions in graphene bilayers

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Graphene, a single sheet of graphite with honeycomb lattice structure, has massless carriers with tunable density and polarity. We investigate the ground state phase diagram of two graphene sheets (embedded in a dielectric) separated by distance d where the top layer has electrons and the bottom layer has holes, using mean-field theory. We find that a uniform excitonic condensate occurs over a large range of carrier densities and is weakly dependent on the relative orientation of the two sheets. We obtain the excitonic gap, quasiparticle energy and the density of states. We show that both, the condensate phase stiffness and the mass of the excitons, with massless particles as constituents, vary as the square-root of the carrier density, and predict that the condensate will not undergo Wigner crystallization.

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Introduction: Over the past three years, graphene has emerged as the unique candidate that provides a realization of two-dimensional massless fermions whose carrier density and polarity are tunable by an external gate voltage [1]. Subsequent experimental and theoretical investigations have led to a thorough re-examination of some of the properties of linearly dispersing massless fermions [2, 3]. The truly two dimensional (2D) nature of graphene permits control and observation of local carrier density and properties [4, 5, 6]. In graphene bilayers, the ability to change the carrier polarity of an individual layer implies that the interlayer Coulomb interaction can be tuned from repulsive to attractive. This raises the possibility of formation of electron-hole bound states or indirect excitons, albeit with massless fermions as its constituents. Properties of such bound states of *massless particles* are an open question; the only other example, to our knowledge, is the proposed color superconductivity in dense quark matter [7]. Graphene bilayers provide an ideal and unique candidate for straightforward experimental investigations of such phenomena.

A uniform Bose-Einstein condensate of excitons in electron-hole bilayers occurs when the interlayer distance is comparable to the distance between the particles within each layer [8, 9]. These excitons have mass $m_{ex} = m_e + m_h$ where m_e (m_h) is the band mass of the electron (hole). At high densities, dipolar repulsion between the excitons can lead to a condensate ground state with broken translational symmetry: a supersolid [10]. Biased bilayer quantum Hall systems near total filling factor one have shown uniform excitonic condensation [11]. In this case, the exciton mass is determined solely by interlayer Coulomb interaction and is independent of the bias voltage [12, 13]. These observations raise the questions: What is the mass of an exciton with massless constituents? Will such an excitonic condensate lead to a supersolid if the dipolar repulsion between such excitons (with a nonzero mass) is increased?

In this paper, we investigate the excitonic condensation in two graphene sheets embedded in a dielectric and separated by a distance $d \gg a$ (a=1.4Å is the honeycomb lattice size) so that the tunneling between the layers is negligible, but interlayer Coulomb interaction is not. The layers have opposite polarity and equal density of carriers n_{2D}. We remind the Reader that in graphene, in the continuum limit, the length-scale $1/k_F$ and the energy-scale E_F are both set by the density of carriers n_{2D} ($k_F = \sqrt{\pi n_{2D}}$ is the Fermi momentum, $E_F = \hbar v_G k_F$ is the Fermi energy, and $v_G \sim c/300$ is the speed of massless carriers). Therefore, the ground state phase diagram depends only on one dimensionless parameter $k_F d$. This is markedly different from conventional bilayer systems parameterized by $(d/a_B, r_s)$ where a_B is the band Bohr radius and $r_s = 1/\sqrt{\pi a_B^2 n_{2D}}$ [9], as well as biased bilayer quantum Hall systems, parameterized by $(d/l_B, \Delta\nu)$ where l_B is the magnetic length and $\Delta\nu$ is the filling factor imbalance [13, 14, 15, 16].

We use the mean-field theory to obtain the ground-state phase diagram as a function of $k_F d$. We find that a) excitonic condensation occurs at all densities as long as $k_F d \sim 1$. b) the condensate properties are weakly sensitive to the relative orientation of the two sheets (stacking). c) the superfluid phase stiffness ρ_s and the exciton mass have a $\sqrt{n_{2D}}$ dependence. d) the excitonic condensate does not undergo Wigner crystallization in spite of dipolar repulsion between excitons with a nonzero mass.

The plan of the paper is as follows. In the next section, we present the mean-field Hamiltonian [17] and briefly sketch the outline of our calculations. In the subsequent section, we show the results for the excitonic gap $\Delta_{\mathbf{k}}$, the quasiparticle energy $E_{\mathbf{k}}$, and the quasiparticle density of states $D(E)$. We discuss the density dependence of the superfluid stiffness ρ_s and the mass of the excitons. In the last section, we show that these results are equivalent to absence of Wigner crystallization, and mention the implications of our results to experiments.

Mean-field Model: We consider two graphene sheets embedded in a dielectric separated by distance d with chem-

ical potentials in the two layers adjusted so that the top layer (denoted by pseudospin $\tau = +1$) has electrons and the bottom layer (denoted by pseudospin $\tau = -1$) has holes with the same density. We consider two stackings: the Bernal stacking that occurs naturally in graphite, and the hexagonal stacking in which each sublattice (A and B) in one layer is on top of the corresponding sublattice in the other layer. Since the Hamiltonian in the continuum description is SU(4) symmetric in the spin and valley indices, we ignore those indices for simplicity. In the continuum limit, the single-particle Hamiltonian for carriers in layer τ is [18]

$$\hat{H}_0 = \Sigma_{\mathbf{k}\alpha}(\alpha\hbar v_G k) c^\dagger_{\mathbf{k}\alpha\tau} c_{\mathbf{k}\alpha\tau} \tag{1}$$

where $\mathbf{k}$ is the momentum measured from the $\mathbf{K}$-point and $\alpha = \pm$ denote the conduction and valance bands that result from diagonalizing the Hamiltonian in the sublattice-basis. $c^\dagger_{\mathbf{k}\alpha\tau}$ ($c_{\mathbf{k}\alpha\tau}$) is creation (annihilation) operator for an electron in band α in layer τ with momentum $\mathbf{k}$. We point out that for the hexagonal stacking, $c^\dagger_{\mathbf{k}\alpha\tau} = [c^\dagger_{\mathbf{k}A\tau} + \alpha e^{-i\theta_\mathbf{k}} c^\dagger_{\mathbf{k}B\tau}]/\sqrt{2}$ is independent of the layer index τ. For the Bernal stacking, the creation operators in the two layers are related by *complex* conjugation, $c^\dagger_{\mathbf{k}\alpha\tau} = [c^\dagger_{\mathbf{k}A\tau} + \alpha e^{-i\tau\theta_\mathbf{k}} c^\dagger_{\mathbf{k}B\tau}]/\sqrt{2}$ where $\theta_\mathbf{k} = \tan^{-1}(k_y/k_x)$. The interaction Hamiltonian consists of intralayer Coulomb repulsion $V_A(\mathbf{q}) = 2\pi e^2/\epsilon q$ and interlayer Coulomb attraction $V_E(\mathbf{q}) = -V_A(\mathbf{q})\exp(-qd)$. ($\epsilon$ is the dielectric constant). Using standard mean-field techniques [17], we obtain the following mean-field Hamiltonian

$$\hat{H} = \sum_\mathbf{k} \left[\begin{array}{cc} e^\dagger_\mathbf{k} & h_{-\mathbf{k}} \end{array}\right] \left[\begin{array}{cc} \epsilon_\mathbf{k} - \mu & \Delta_\mathbf{k} \\ \Delta^*_\mathbf{k} & -\epsilon_\mathbf{k} + \mu \end{array}\right] \left[\begin{array}{c} e_\mathbf{k} \\ h^\dagger_{-\mathbf{k}} \end{array}\right]. \tag{2}$$

Here $e^\dagger_\mathbf{k} = c^\dagger_{\mathbf{k}++}$ creates an electron in the conduction band ($\alpha = +$) in the top layer ($\tau = +1$) and $h^\dagger_{-\mathbf{k}} = c_{\mathbf{k}--}$ creates a *hole* in the valance band ($\alpha = -$) in the bottom layer ($\tau = -$). The term $\epsilon_\mathbf{k}$ contains single-particle energy, capacitive Hartree self-energy and the intralayer exchange self-energy. The off-diagonal term $\Delta_\mathbf{k}$ is proportional to the excitonic condensate order parameter $\langle h_{-\mathbf{k}} e_\mathbf{k}\rangle$. The eigenvalues of the mean-field Hamiltonian are given by $\pm E_\mathbf{k} = \pm\sqrt{(\epsilon_\mathbf{k} - \mu)^2 + \Delta^2_\mathbf{k}}$. We consider mean-field states with a real $\Delta_\mathbf{k} = \Delta^*_\mathbf{k}$, and spatially uniform density. It is straightforward to diagonalize the Hamiltonian and obtain the mean-field equations [9]

$$\epsilon_\mathbf{k} = \hbar v_G k + \frac{e^2 n_{2D}}{C} - \frac{1}{2}\int_{\mathbf{k}'} V_A(\mathbf{k}-\mathbf{k}')\left[1 - \frac{\xi'_\mathbf{k}}{E'_\mathbf{k}}\right] \tag{3}$$

$$\Delta_\mathbf{k} = -\frac{1}{2}\int_{\mathbf{k}'} V_E(\mathbf{k}-\mathbf{k}') f(\theta_{\mathbf{k},\mathbf{k}'}) \frac{\Delta_{\mathbf{k}'}}{E'_\mathbf{k}} \tag{4}$$

where $\xi_\mathbf{k} = \epsilon_\mathbf{k} - \mu$, $C = \epsilon/2\pi d$ is the capacitance per unit area, and $\theta_{\mathbf{k},\mathbf{k}'} = \theta_\mathbf{k} - \theta_{\mathbf{k}'}$. The form factor for the two stackings are

$$f(\theta_{\mathbf{k},\mathbf{k}'}) = \left\{\begin{array}{cc} (1+\cos\theta_{\mathbf{k},\mathbf{k}'}) & \text{Hexagonal} \\ \cos\theta_{\mathbf{k},\mathbf{k}'}(1+\cos\theta_{\mathbf{k},\mathbf{k}'}) & \text{Bernal} \end{array}\right. . \tag{5}$$

We point out that the self-energy in Eq.(3) takes into account both intrinsic and extrinsic contributions that cancel the $\cos\theta_{\mathbf{k},\mathbf{k}'}$-dependent terms in the form factor and make the results independent of the ultra-violet cutoff [19, 20]. Therefore the intra-layer self-energy in Eq.(3) is the same as that for a conventional system [19, 20]. The chemical potential μ is determined by the carrier density that takes into account the four-fold spin and valley degeneracy

$$n_{2D} = 4\int_\mathbf{k}\left[1 - \frac{\xi_\mathbf{k}}{E_\mathbf{k}}\right]. \tag{6}$$

It is straightforward to derive similar equations for a conventional electron-hole system [9]. They are obtained by changing the single-particle dispersion to a quadratic and replacing the form factor $f(\theta_{\mathbf{k},\mathbf{k}'})$ by a constant $f = 2$. We solve Eqs. (3), (4) and (6) iteratively to obtain self-consistent results.

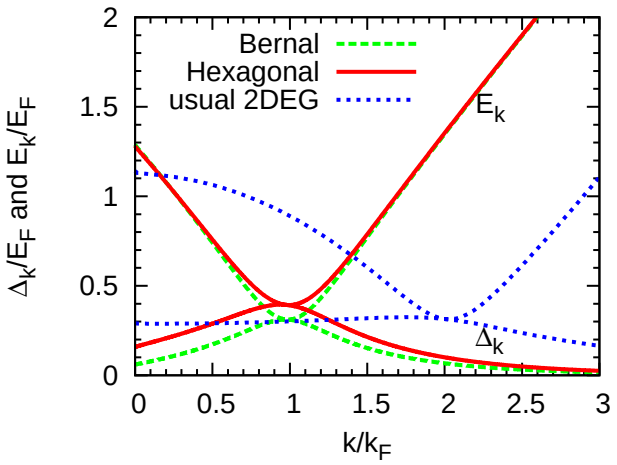

FIG. 1: (Color online) Excitonic gap $\Delta_\mathbf{k}$ and the quasiparticle energy $E_\mathbf{k}$ in graphene bilayer for Bernal (green dashed) and hexagonal (red solid) stacking with $k_F d = 1$. The quasiparticle spectrum $E_\mathbf{k}$ becomes linear with a renormalized velocity $\tilde{v}_G > v_G$ for large $k \gg k_F$. The dotted blue curves show corresponding results for an electron-hole system at $r_s = 2.7$ and $k_F d = 1$ when plotted using relevant (atomic) unit for energy [9].

Results: Figure 1 shows the excitonic gap $\Delta_\mathbf{k}$ and the quasiparticle energy $E_\mathbf{k}$ for the Bernal (green dashed) and the hexagonal (red solid) stacking. The excitonic gap $\Delta_\mathbf{k}$ is maximum at the Fermi momentum k_F where the quasiparticle energy $E_\mathbf{k}$ is minimum. Since the electron-hole Coulomb interaction is always attractive, the excitonic condensate order parameter is nonzero down to the bottom of the Fermi sea, $\Delta_{\mathbf{k}=0} \neq 0$. Our results predict that the hexagonal-stacked system will have a larger excitonic gap than the Bernal-stacked system.The quasiparticle energy $E_\mathbf{k}$ becomes linear at large $k \gg k_F$, since the constituent particles of the exciton have a linear dispersion. The speed of these quasiparticles is increased due

to intralayer exchange self-energy [20, 21, 22] although the increase is modest, $\sim 10\%$. Corresponding results for a conventional electron-hole system (blue dotted) are also shown in Fig. 1.

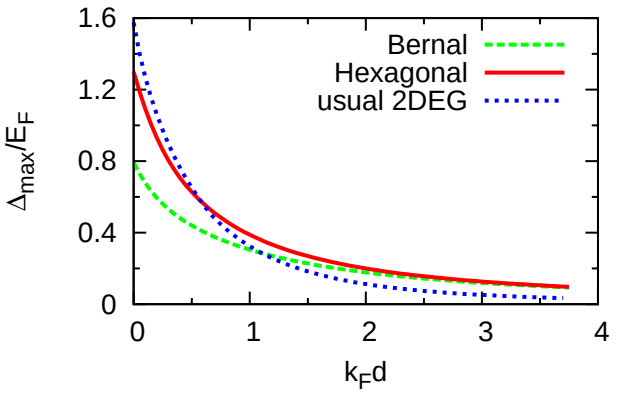

FIG. 2: (Color online) Dependence of the graphene bilayer excitonic gap $\Delta_m = \mathrm{Max}(\Delta_{\mathbf{k}})$ on interlayer distance d for Bernal (green dashed) and hexagonal (red solid) stacking. This gap can be tuned by changing n_{2D} for a given sample. Corresponding result for a conventional system at $r_s = 2.7$ is shown in dotted blue.

Figure 2 shows the dependence of the maximum excitonic gap Δ_m on k_Fd. We find that Δ_m is weakly dependent on the stacking and decays rapidly when $k_Fd \gg 1$. *This result implies that the excitonic condensation is a robust phenomenon that will not require precise alignment of the two graphene sheets when they are being embedded in a dielectric.* With typical graphene carrier densities $n_{2D} \sim 10^{12}/\mathrm{cm}^2$ and $d \sim 100$Å or $k_Fd \sim 1$, the excitonic gap is appreciable, $\Delta_m \sim 30$ meV.

A direct probe of the excitonic gap is the quasiparticle density of states. For graphene with no interactions, the density of states is linear, $D_0(E) = 2E/\pi\hbar^2v_G^2$. In the excitonic condensate phase, for intermediate energies $\Delta_m \le E \le E_{\mathbf{k}=0}$ there are two rings in the phase-space consistent with that energy: one with $k < k_F$ and the other with $k > k_F$. Therefore the quasiparticle density of states is given by $D(E) = D_<(E) + D_>(E)$ where $D_<$ ($D_>$) denotes the density of states from respective rings. Figure 3 shows $D_<(E)$ (green dashed) and $D_>(E)$ (red solid); they are both zero for $E < \Delta_m$ and diverge at Δ_m as is expected. Note that $D_<(E) = 0$ for $E > E_{\mathbf{k}=0}$, since there are no states for $k < k_F$ with energies higher than $E_{\mathbf{k}=0}$. The asymmetry in $D_<$ and $D_>$ for $E \gg \Delta_m$ is due to the linear dispersion of carriers and the nonzero electron-hole pairing that extends to the bottom of the Fermi sea, $\Delta_{\mathbf{k}=0} \neq 0$ [23]. The inset shows corresponding results a conventional system, where the density of states without interactions is constant, $D_0(E) = m/\pi\hbar^2$.

Superfluidity of a uniform Bose-Einstein condensate is characterized by a non-zero phase stiffness ρ_s that quantifies the energy of a condensate with a linearly winding phase, $E(Q) = \rho_sQ^2A/2$ where A is the area of the sample and the phase of the condensate varies as $\Phi(x) = Qx$. For graphene, since E_F is the sole energy scale (at zero temperature), it follows from dimensional analysis that phase stiffness must scale linearly with the Fermi energy, $\rho_s = g(k_Fd)E_F$ where $g(x)$ is a dimensionless function that satisfies $g \sim O(1)$ [24] when $0 \le x \lesssim 1$ and $g \to 0$ for $x \gg 1$. Hence, the phase stiffness is given by $\rho_s = g(k_Fd)\hbar v_G\sqrt{\pi n_{2D}}$. The condensate energy $E(Q)$ can also be expressed, in the particle-picture, as the kinetic energy of excitons that have condensed in a state with center-of-mass momentum $\hbar Q$. Thus, $E(Q) = N\hbar^2Q^2/2m_{ex}$ where m_{ex} (N) is the mass (number) of condensed excitons [25]. Equating the two expressions for energy implies $m_{ex} = n_{2D}\hbar^2/\rho_s \propto \sqrt{n_{2D}}$. Thus *we predict that the phase stiffness ρ_s and the exciton mass will both vary as the square root of the carrier density.* We emphasize that these results are unique to graphene and, as we will show in the next section, are *equivalent* to the absence of excitonic Wigner crystallization in graphene bilayers [10, 26].

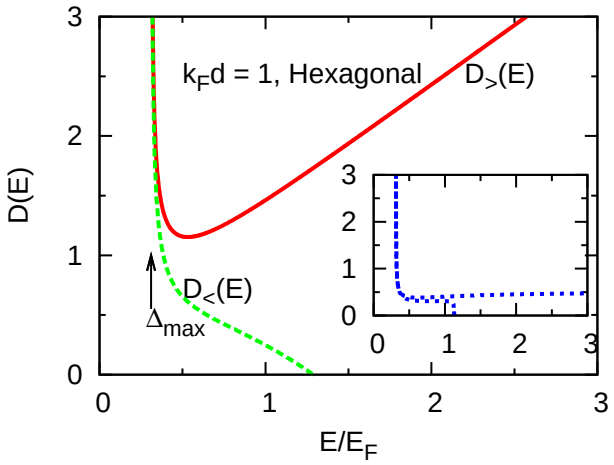

FIG. 3: (Color online)Quasiparticle density of states contributions $D_<(E)$ (green dashed) associated with states with $k \le k_F$, and $D_>(E)$ (red solid) associated with states $k \ge k_F$. These results are for hexagonal stacked graphene bilayers with $k_Fd = 1$. Both diverge at $E = \Delta_m$, as expected. The total density of states $D = D_< + D_>$ can be probed by differential conductance for tunneling from a metal into the condensate. The inset shows corresponding results for an electron-hole system at $r_s = 2.7$ and $k_Fd = 1$. All results are expressed in their respective units.

Discussion: In this paper, we have investigated the properties of excitonic condensates in graphene bilayers. Our calculations predict that excitonic condensation will occur at all carrier densities as long as $k_Fd \sim 1$, and that the strength of the condensate, as measured by the excitonic gap Δ_m is relatively insensitive to the stacking.

The mean-field results presented in this paper are obtained at zero temperature T=0. (Finite temperature

analysis gives a critical temperature $T_{MF}/E_F \sim 0.2$ or $T_{MF} \sim 20$ meV. This is an artifact of the mean-field approximation.) In two dimensions, the critical temperature T_c for Bose-Einstein condensation is zero, but the superfluid properties survive for $T \leq T_{KT}$ where T_{KT} is the Kosterlitz-Thouless transition temperature. Therefore, our results will be valid at nonzero temperature $T \ll T_{KT}$ [27]. A weak disorder will suppress the excitonic condensate order parameter and reduce the excitonic gap, an effect equivalent to increasing the value of $k_F d$. Therefore, we have ignored the effects of a weak disorder potential.

In our analysis, we have only considered excitonic condensation with uniform density. In conventional (quantum Hall electron-hole) bilayers, varying d and r_s (ν) leads to excitonic condensates with lattice structure [10, 26, 28]. The origin of the lattice structure is Wigner crystallization of carriers in an isolated layer at large r_s (small ν). Graphene does not undergo Wigner crystallization as its carrier density is changed [29]. Therefore, we expect that the excitonic condensate in graphene bilayers remains uniform. Now we show that this result is equivalent to our predictions for density dependence of ρ_s and m_{ex}. The quantum kinetic energy of an exciton, associated with localizing it within a distance $1/k_F$, is $K = \hbar^2 k_F^2/2m_{ex}$. The potential energy due to the dipolar repulsion between them is $P = e^2 d^2 k_F^3/\epsilon$. Hence their ratio is given by $P/K = e^2 d^2 k_F m_{ex}/\epsilon\hbar^2$. Wigner crystallization occurs when the ratio $P/K \gg 1$. This ratio will solely be a function of $k_F d$ - no matter what the value of d is - *if and only if* $m_{ex} \propto k_F = \sqrt{\pi n_{2D}}$. Therefore, results in the last section show that *the excitonic condensate in graphene will not undergo Wigner crystallization* in spite of the dipolar repulsion between excitons with a quadratic dispersion. This result, too, is unique to graphene and is markedly different from the behavior of dipolar excitonic condensates in conventional bilayers. It is interesting that the mass of these effective bosons has the same density dependence and order of magnitude as the cyclotron mass of fermionic carriers in graphene [1].

The onset of excitonic condensation can be detected by a divergent interlayer drag [30]. A uniform in-plane magnetic field $B_{||}$ between the two graphene sheets is expected to induce a (counterflow) supercurrent J_d in such a condensate [31], $J_d = 2\rho_s e^2 d B_{||}/\hbar^2$. The phase stiffness ρ_s and its density dependence can be directly obtained from experimental measurements of the counterflow supercurrent. The verification (or falsification) of our predictions, including the density dependence of ρ_s and m_{ex}, will deepen our understanding of properties and condensation of excitons with massless fermions as constituent particles.

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[23] In N-dimensions, fermions with dispersion $\epsilon_k \sim k^\alpha$ and excitonic gap Δ will have $D_{<(>)}(E) = D_0[\mu \mp \sqrt{E^2 - \Delta^2}]^{N/\alpha - 1}$ for $\Delta \leq E \leq \sqrt{\mu^2 + \Delta^2}$, as k reduces from k_F to 0. Thus, for $N = \alpha$, $D_<(E) = D_>(E) = D_0$ where $D_0 = E/\sqrt{E^2 - \Delta^2}$.
[24] $T = 0$ phase stiffness, calculated in Ref. [27] via mean-field analysis reproduces the same functional form and implies that $g = 1/4\pi$.
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